Mixed Organic/ Inorganic Aerosols at High Relative Humidities

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Project Background

- Aerosols have much greater light scattering, and potentially greater light absorption, when they are hydrated.
- Light scattering of inorganic, certain organic and mixed aerosols as a function of Relative Humidity (RH) has been studied in the past, but the RH in these studies is typically < 85%.
- Light absorption of organic and mixed organic/ inorganic aerosols has rarely been examined at high RH because most light absorption measurement techniques are not applicable for such conditions.
- The RH range between 85 and 95% is important because aerosol diameter and light scattering can change rapidly as RH increases. The range also represents the transition between haze aerosol and clouds (transition zone).
- For radiative-transfer modeling, a typical approach is to assume that observations taken < 85% RH constrain the functional form and magnitude of the aerosol scattering response. The functional form of the growth is assumed to hold at RH > 85%, but this portion of the response is not well constrained with measurements.
- The water uptake of aerosols depends on the aerosol chemical composition in particular on the mass ratio of organic and inorganic substances. (Figure1)

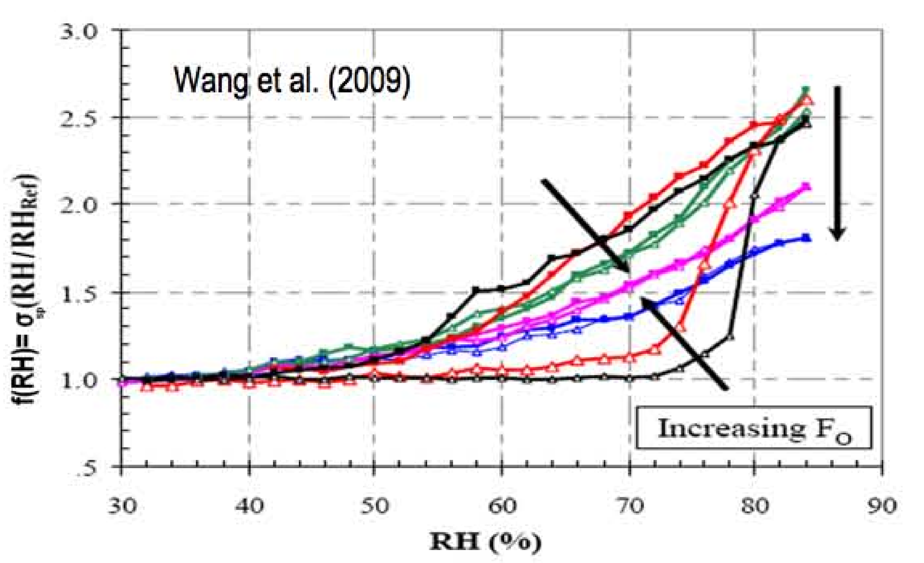


Figure 1. Humidograms for aerosol mixtures containing $(NH_4)_2SO_4$, succinic acid and malonic acid at select F_O values. F_O represents the mass ratio of organic (equal mass of succinic and malonic acids) to $(NH_4)_2SO_4$. f(RH) represents the ratio of scattering at a particular RH to dry scattering.

Goals

- Measure the optical properties (scattering and absorption) of mixed organic/ inorganic aerosols with measurements at a RH range of 85 - 95% with laboratory measurements.
- Develop a parameterization for future radiative-transfer model based on the measured results.

Objectives

1. Assemble analytical equipment to measure light scattering, backscattering and absorption by aerosols at high sub-saturated RHs and at multiple visible wavelengths.
2. Measure these optical properties for specific organic aerosol fractions, namely oxygenated aromatic species and polymerized light-absorbing aerosol, both pure and mixed with inorganic solute.
3. Model the optical properties of the aerosol independent of the optical measurements and evaluate closure between measured and predicted optical properties.
4. Identify chemical measurements that correspond to fractions of organic carbon with distinct climate-relevant properties.
5. Provide emission- and process-linked parameterizations of optical properties in the form required for implementation in next-generation climate models.

Approach

I. Development of the measurement equipment for high RHs (Objective 1)

We propose to use the difference between light extinction and light scattering to determine the light absorption of aerosols.

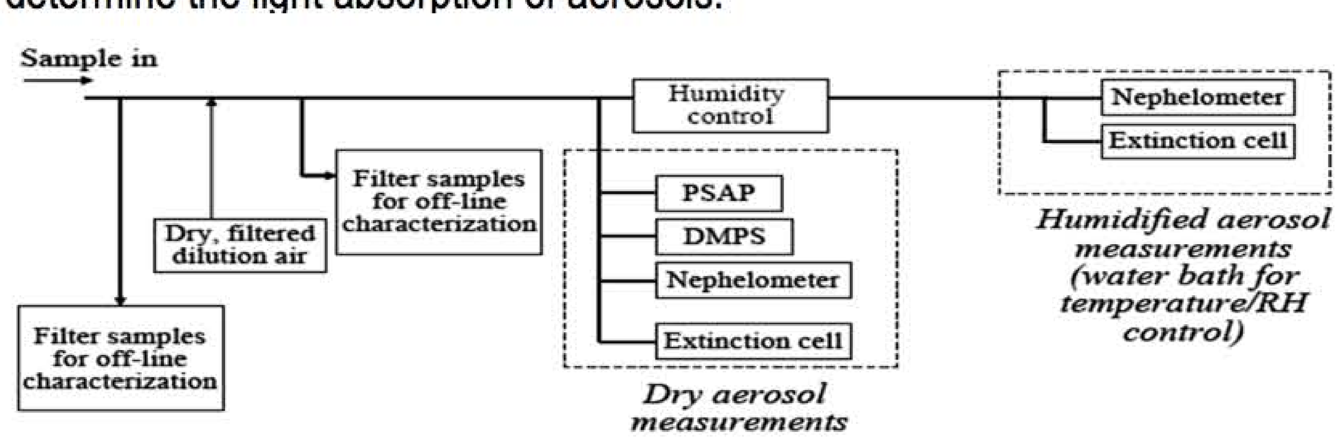


Figure 2. Schematic of measurement system for characterizing optical and chemical properties of aerosol.

II. Laboratory tests (Objective 2)

The main focus of the study is Organic Carbon (OC) aerosols generated from controlled biomass combustion. (Table 1)

Table 1. Proposed test aerosols

Test Group	Solute	Processing/ Coating	Purpose of tests
1	Ammonium Sulfate	None	System benchmark
	Glutaric acid	None	System benchmark
	Succinic Acid	None	System benchmark
2	Humic acid standard	None	Compare with previous measurements
	Humic acid standard	Ammonium sulfate	Compare with previous measurements
3	OC from biomass, low T (oxygenated)	None	Examine optical properties and hygroscopic response of this "new" material
	OC from biomass, high T (polymerized)	None	"
4	OC from biomass, low & high T	Ammonium sulfate	Determine behavior of mixed aerosol
5	OC from biomass, low & high T	Ammonium sulfate/long residence time	Test whether catalyzed reactions affect optical properties and hygroscopic growth
6	OC from biomass, low & high T	Ammonium nitrate	Determine behavior of mixed aerosol
7	OC from biomass, low & high T	Ammonium nitrate/long residence	Test whether reactions affect optical properties and hygroscopic growth

III. Closure studies and model connections (Objectives 3, 4 and 5)

- Closure will be evaluated for optical properties (Mie model) and for aerosol water uptake (ZSR model).
- The parameterization of the results will focus on identifying fractions of the atmospheric aerosol that are measurable in field studies, predictable in models, and are linked with specific, measured climate-relevant properties.

Accomplishments: Improvement and Benchmarking of the Extinction Cell

The black box section of the Short Path Extinction Cell (SPEC) was redesigned to maintain the optical alignment, reduce signal reflections, reduce electrical noise, and improve signal stability. (Figure 4 and Figure 5)

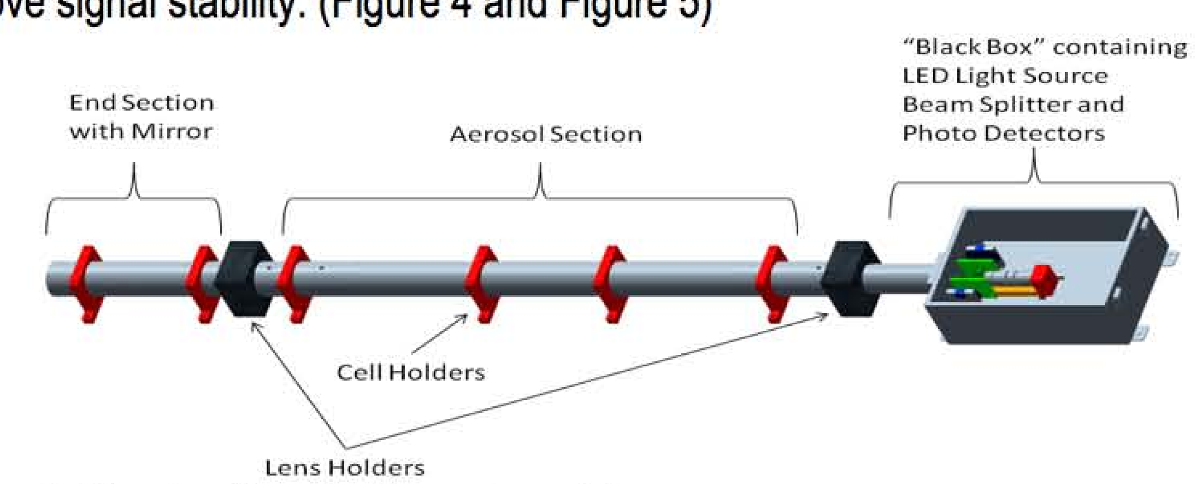


Figure 4. Short path extinction cell model.

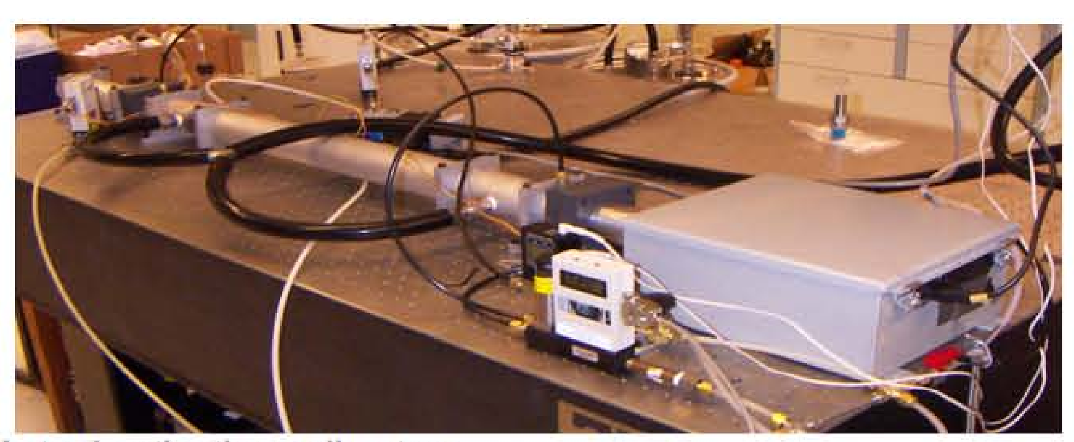


Figure 5. Actual extinction cell set-up.

Benchmark tests were performed by running tests with white $(NH_4)_2SO_4$ aerosol for which light scattering should match light extinction. The light scattering was measured with a TSI 3563 nephelometer. (Figure 6)

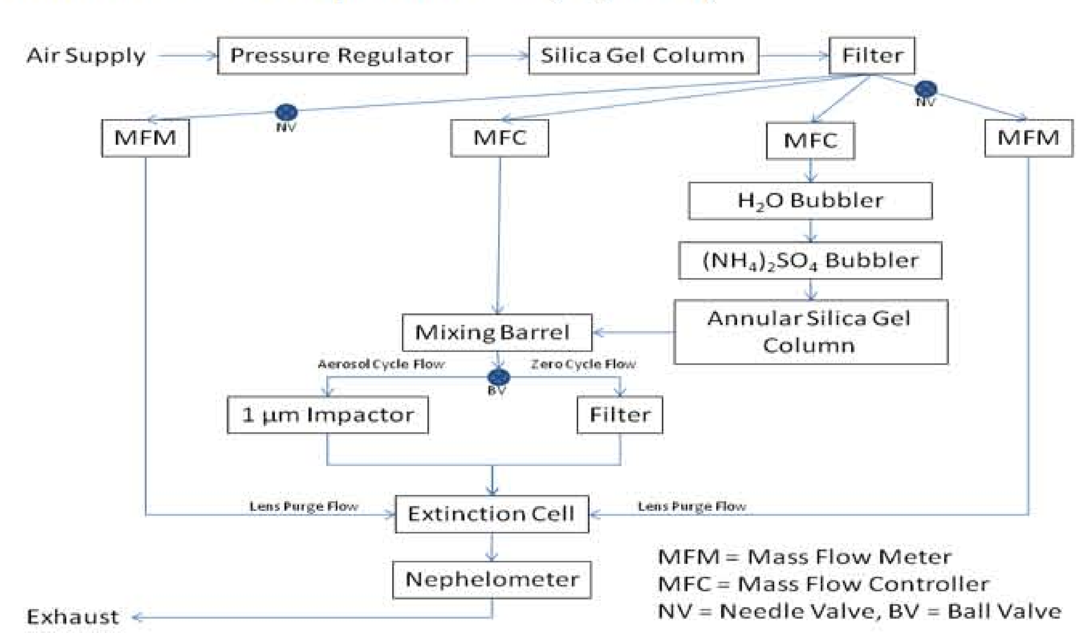


Figure 6. Schematic of the benchmarking set-up used. The SPEC generates its signal in a two-cycle mode by comparing the signal reference ratio for clean air and for aerosols, respectively.

Results

The redesign resulted in a better matching of the SPEC and nephelometer signals. The noise level of the SPEC has also been reduced (Figures 7 and 8).

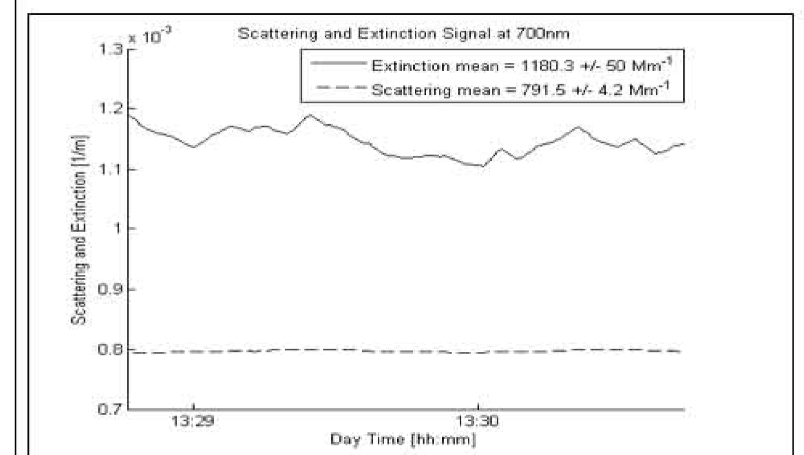


Figure 7. Red (700 nm) signals for $(NH_4)_2SO_4$ aerosols at a scattering level of 800 Mm^{-1} before extinction cell redesign.

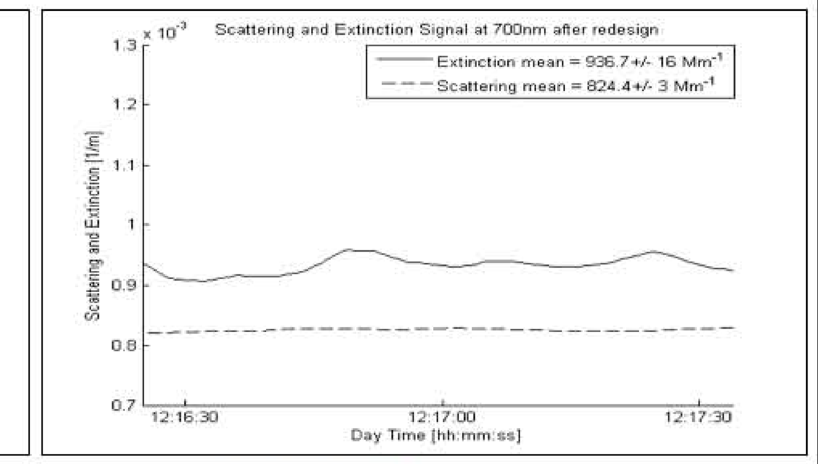


Figure 8. Red (700 nm) signals for $(NH_4)_2SO_4$ aerosols at a scattering level of 800 Mm^{-1} after extinction cell redesign.

The average ratio between scattering and extinction shows a constant pattern at all three wavelengths after the redesign. (Figures 9 and 10)

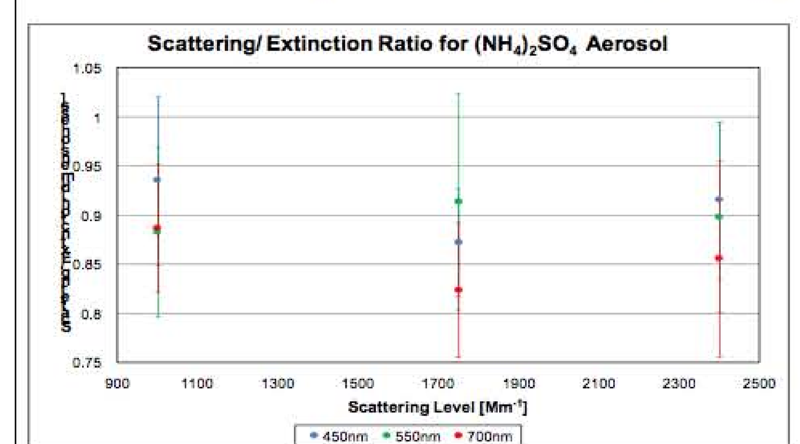


Figure 9. Average ratio of scattering and extinction for white aerosols before extinction cell redesign.

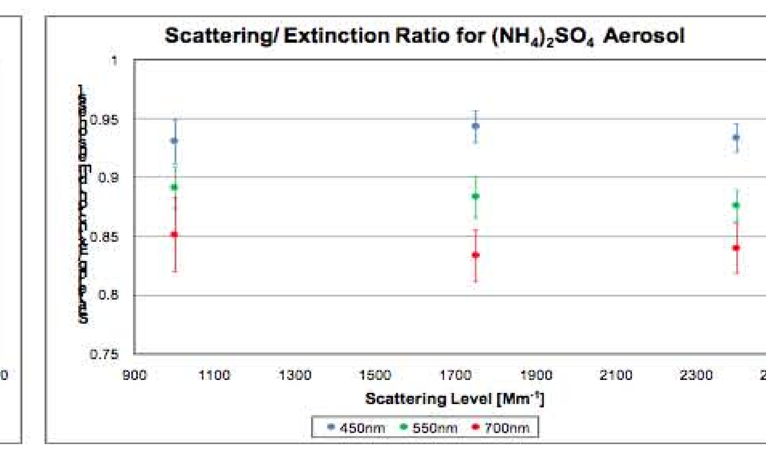


Figure 10. Average ratio of scattering and extinction for white aerosols after extinction cell redesign.

Current Issues

Current work addresses the temperature dependency of the signal and reference photo detectors to improve the long term signal stability. (Figures 11 and 12)

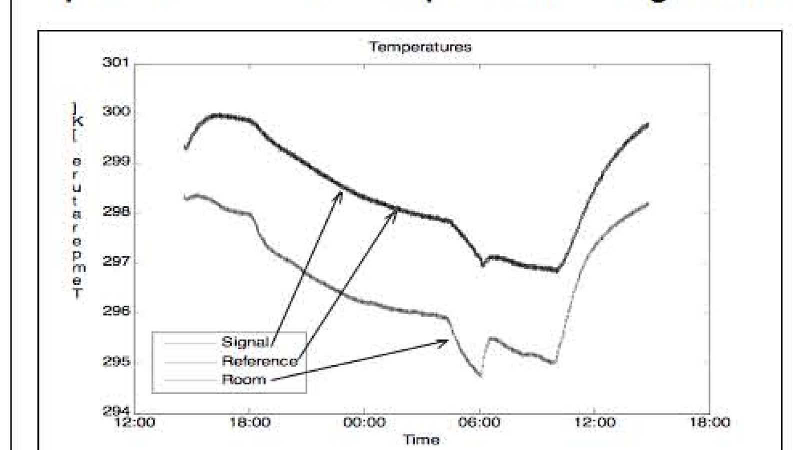


Figure 11. Temperature profiles.

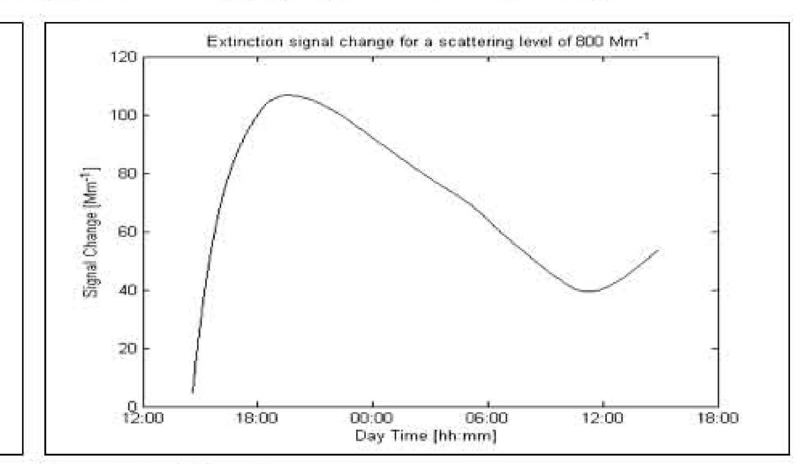
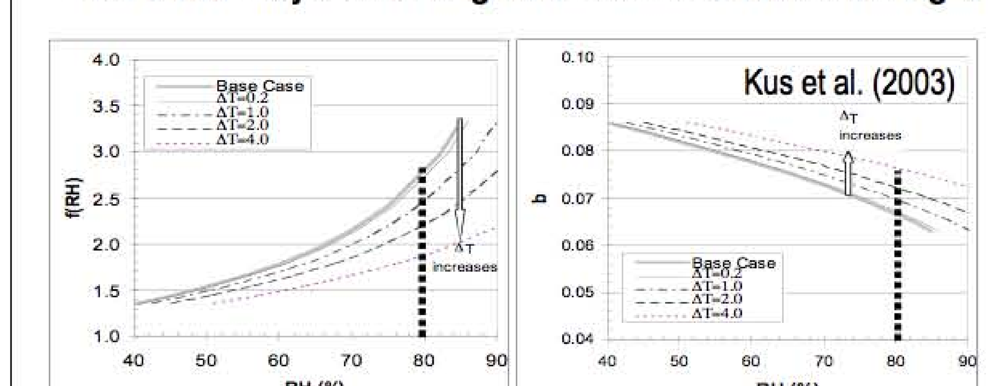


Figure 12. Signal response.

Future Challenges

The three major challenges that will be faced during the project duration:



1. Determination of light absorption with difference method.
2. High RH control, especially the prevention of instrument heating (Figures 13 and 14)
3. Constant aerosol generation from biomass combustion.

Figures 13. and 14. Influence of temperature on growth factor $f(RH)=(d_p(RH)/d_p(dry))$ and backscatter ratio $b=(\sigma_{bsp}/\sigma_{sp})$.

Acknowledgements

Optical Properties of Moderately-Absorbing Organic and Mixed Organic/ Inorganic Particles at Very High Humidities
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